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THE CORROSIVE ACTIVITY OF RUBBERS AND COMPOUNDS BASED ON THEM (--ETC(U)

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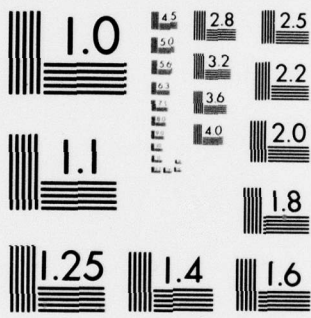
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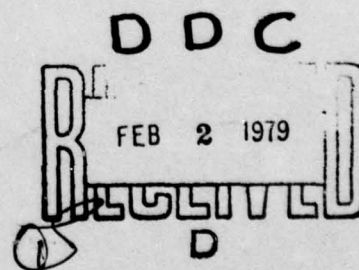
by

A.K. Smolich

A.L. Labutin

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THE CORROSIVE ACTIVITY OF RUBBERS AND COMPOUNDS BASED ON THEM
(KORROZIONNAYA AKTIVNOST KAUCHUKOV I KOMPOZITSII NA IKH OSNOVE)

by

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A. K. Smolich
A. L. Labutin

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Translator

J.W. Palmer.

(USSR)

Translation editor

E. Kay

EDITOR'S SUMMARY

The author reviews the literature published during the last 25 years on the corrosion of metals in contact with rubber, its causes, assessment and means of combating it.

Metals can be attacked by, (1) impurities such as catalyst residues from polymerisation, (2) the products of chemical breakdown of the polymer during processing, subsequent storage, or in service or (3) compounding ingredients, antioxidants, fillers etc.

Tests to predict 'in service' behaviour are usually made at high humidity and elevated temperatures. Various methods are used to assess the results.

Means of reducing corrosion include purification of the base polymer, the inclusion of acid acceptors in the vulcanisate, post cure to remove harmful volatiles, applying a protective coating to the metal and control of environmental conditions.

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NOTES:-

- SKF-26 (SKF - fluorine containing rubbers)
- SKB rubber (sodium-catalysed polybutadiene rubber based on butadiene synthesised from alcohol; \equiv 'Buna' rubber)
- SKN-18 (copolymers of butadiene and acrylonitrile obtained in an aqueous emulsion). SKN-18 (acrylonitrile content: 17-20%)
- SKN-26 Acrylonitrile content: 27-30%
- SKF-32 (SKF fluorine - containing rubbers)
- SKMS-30 (SKMS - copolymers of butadiene and α -methylstyrene obtained by emulsion polymerisation at 48-50°C). Figure denotes per cent of methylstyrene)
- SKT (heat-resistant polydimethyl siloxane (silicone) rubber)
- Y30-M

Information about the corrosion of metals during contact with certain rubbers and sealants has been available in the literature for a long time (1-6). Particular attention has been devoted to this problem in recent years in connection with the extension of the use of polymer materials in various branches of technology and, in particular, in electronic instrument making. It is well known that the corrosion of metals in contact with polymer materials may be the cause of failure of instruments or other appliances⁷, as well as of seal failure of the system. In certain conditions even such resistant metals as gold may be subjected to corrosion, for example during contact with materials based on chloroprene rubber^{8,9}.

The surface attack of metals which are in contact with rubber, or a sealant, may be caused by the corrosion activity both of the polymer and its technological admixtures, and of the vulcanising agents, anti-oxidants and other ingredients. Increased humidity and temperature and reduced atmospheric pressure, act as stimulating factors. In those cases where the atmosphere, in which the rubber-metal components are operating, is contaminated by corrosive gases, *eg* CO_2 , NH_3 , H_2S , the role of the rubber in the corrosive attack of the metal cannot by any means always be specifically determined.

Synthetic rubbers with a hydrocarbon structure, like natural rubber, are inert in a corrosion sense in ordinary conditions, but may become corrosive when their chemical stability is destroyed as a result of thermal or mechanical action. In these conditions when the macro-molecules are attacked they form highly active free radicals which enter into a chemical reaction with the surface of the metal in contact with the polymer¹⁰. In real conditions of operation the stability of the polar rubbers is frequently reduced, not merely by thermal and mechanical action, but also by natural ageing. Thus chloroprene rubbers (Nairites, Neoprenes) after lengthy operation in an air medium with 100% relative humidity in contact with copper and its alloys, even at 60°C show marked corrosive activity^{1,11,12}. Hydrogen chloride is the corrosive agent in this case, while at temperatures of the order of 120°C , chlorine also is involved^{8,13,14}.

The very aggressive hydrogen fluoride which is separated as a result of their partial thermal breakdown at $200-250^\circ\text{C}$ ^{15,16,17} is responsible for corrosive activity of fluorine-containing rubbers of the SKF-26 type. The corrosion of metals in contact with acrylate and nitrile rubbers occurs due to the formation of acrylic and formic acids^{12,17}, and in the case of contact with polyurethanes it occurs as a result of the separation of ammonia and hydrogen

cyanide^{12,18}. Certain low molecular weight liquid rubbers, particularly with terminal functional groups show a low corrosive activity.

Technological admixtures of industrial rubbers (residues of catalysts, emulsifiers, traces of metals and so on) may have corrosive activity or play the part of catalysts^{8,11,19,20}. Thus, the alkali reaction of an aqueous extract of SKB rubber provides evidence of the presence of a residue of a catalyst in it. Naturally, by this, means the rubber when in contact with aluminium which is unstable in an alkali medium, causes its corrosion. Low molecular weight mercaptans and other volatile sulphur compounds which are contained in liquid thiokols, cause rapid darkening of silver, copper and their alloys^{4;21}, p.16.

A number of reports²²⁻³⁵ has been devoted to the corrosive activity of ingredients of rubber mixes; anti-oxidants, vulcanising agents, vulcanisation accelerators, plasticizers and fillers. The majority of anti-oxidants are substituted aromatic amines or derivatives of phenols and naphthols. It is well-known that amines and phenols cause considerable corrosion of copper and its alloys³⁶. Therefore their substituted forms may also have a corrosive action on these metals, although to a lesser degree. At the same time, it is noted²² that certain anti-oxidants of the Agerite White type (N,N'-di-B-naphthyl-p-phenylene diamine) reduce the corrosive activity of butadiene nitrile rubbers with respect to steel.

The majority of investigators is inclined to consider that of all the ingredients introduced into rubber mixes, sulphur has the most unfavourable effect on metals which are in contact with the rubber^{5,11,37}. Butadiene nitrile rubbers of the SKN-18, SKN-26 type vulcanised by sulphur plus thiuram have a high corrosive activity with respect to carbon steel. It is possible to reduce the corrosive activity of these rubbers by the introduction of an inhibitor-mercapto benzimidazole³⁸. In some reports it is pointed out that during contact with rubber which contains sulphur even stainless steel corrodes in the presence of moisture^{39,40}, while an increase in the dosage of sulphur from 1 to 2 part by weight does not accelerate the process of corrosion²⁵. However, with lesser concentrations of sulphur in the rubber, the rate of corrosion starts to decline somewhat. Vulcanisates of butadiene nitrile rubbers with vulcanising systems which contain sulphur, Altax, tetra methyl thiuram monosulphide and tetra methyl thiuram disulphide show a corrosive activity with respect to carbon steel. The following are recommended as the least corrosive vulcanising systems, in parts by weight:

- (1) Sulphur 1.5
plus
tetramethyl thiuram monosulphide - 4.5;
- (2) tetramethyl thiuram disulphide - 5;
- (3) Sulphur - 1.5
plus
tetramethyl thiuram disulphide - 3²²

The introduction of stearic acid in a quantity of 0.5 to 2.0 parts by weight somewhat reduces the corrosive action of butadiene nitrile rubbers. Zinc oxide, the properties of which depend on the method of preparation and the method of processing also has an effect on the corrosive action of rubbers. Replacement of zinc oxide by lead oxide in the rubber mix considerably reduces the rate of corrosion of steel²⁵. Certain vulcanising agents of other special rubbers are more aggressive than sulphur. Thus 101 K resin and ferric chloride; hexamethylene diamine acetate and sulphur; stearic acid, Altax and triethylene tetramine are the most corrosive vulcanising systems for the acrylate elastomers. Elastomers containing peroxides and oxides of metals have a low corrosive action^{17,41}. Fluoro-carbon rubbers with an amine vulcanising system and fluorosilicone rubbers with a peroxide system caused negligible corrosion of steel at 40°C and 100% humidity^{42,43}.

At temperatures above 200°C, the corrosion of steel becomes marked owing to the partial degradation of the polymer, with the evolution of aggressive gases¹⁵. In a number of cases the vulcanising agents have a powerful corrosive action solely in the process of vulcanisation of the mixes. Thus during the curing of rubbers based on low molecular weight siloxanes the K-10s catalyst which produces aggressive acetic acid^{44, p.162} has high corrosive action as regards many metals.

According to the published data, plasticizers of rubber mixes - dibutyl phthalate and dioctyl phthalate cause corrosion^{18,22,25,45}, while the esters of sebacic acid reduce the corrosive action of butadiene nitrile rubbers²².

A number of papers^{11,22,25,41,42} is devoted to the effect of fillers on the corrosive action of rubbers. Since corrosion in the system rubber/metal in air in ordinary conditions occurs as a result of electro-chemical processes it is of great interest to determine the role in these processes of conducting fillers such as carbon black and graphite which have a higher oxidising potential compared with metals. It is observed in the literature that natural rubber, chloroprene, butadiene nitrile and other rubbers filled with carbon black may

cause more powerful corrosion of steel and copper than non-filled rubber^{11,22}. The unequal corrosive action of rubbers containing various brands of carbon black has been shown²². For example, vulcanisates of butadiene nitrile rubbers filled with Vulcan C or Sterling L and S blacks show high corrosive activity, whereas rubbers containing Regal 300 and SRF blacks and even SF graphite have a weak corrosive action. Mineral fillers influence the corrosive activity of butadiene nitrile rubbers in different ways. Rubbers containing pure clays cause the minimum corrosion of steel, rubbers containing calcium carbonate are more active, while calcium silicate and silicon dioxide cause even greater attack on metals²². The corrosive action of fluorocarbon rubbers containing barium sulphate was more powerful than those filled with carbon black⁴².

It follows from the published data that so far it has not been possible to determine any general regularities in the corrosive behaviour of the rubber/metal system. Therefore when developing mixes for rubbers intended for operating in contact with metals, it is necessary to determine the corrosive activity of the finished compound.

The methods of investigation and evaluation of the corrosive activity of rubbers are not standardised, although they are not marked by diversity. Most frequently the tests of specimens of rubber/metal, rubber/metal/rubber or metal/rubber/metal are made in air at high humidity and at varying temperatures. Foreign investigators^{15,46,47} when studying the corrosive activity of acrylic rubbers, butadiene nitrile, and other rubbers placed the test specimens between two steel plates and kept them for only 4 days at 38°C in air at 100% relative humidity. The degree of corrosion was determined visually by means of a scale formed by a photographed collection of metal plates with varying depths of corrosion. The corrosion resistance of the metals is also estimated by means of a 5 point scale⁴¹: 0 - absence of corrosion; 0.5 - very, very slight corrosion; 1.0 - very slight corrosion; 2.0 - slight corrosion; 3.0 - medium corrosion; 4.0 - corrosion; 5.0 - excessive corrosion^{25,41}.

One of the laboratories of General Motors has issued information in which the scale of estimates and the methods of studying the corrosion activity of the acrylate and butadiene nitrile rubbers are given^{41,48}. Many foreign investigators who use the 5 point scale for estimating the corrosion resistance of metals consider it suitable for the use of rubber causing negligible corrosion of metal, amounting to 0.5 to 1.0 points^{22,25,41,49-51}. There is also an accelerated method for estimating the corrosive activity of rubbers, according to which the specimens are aged in air with a relative humidity of 98% at 70°C for 30 days with a check test of the specimens after 15 days³⁸.

In order to determine the corrosive activity of sealants²¹ and compounds¹⁹, made up of liquid or paste-like materials, metal plates with an elastomer coating, cured according to methods indicated in Technical Specifications are tested in moist air. At the same time specimens with a coating on one side of a metal plate are frequently collected into a 'battery' while specimens covered with a sealant or compound on all sides, are usually tested separately. As a standard for the specimens collected in the 'battery', use was made of a collection of the corresponding metal plates between which were laid mica, glass, fluoroplas or other non-electroconducting materials characterised by complete inertness under the test conditions.

In some cases rubber-metal specimens are tested in sea and fresh water, solution of salts or other corrosive media for a long period⁵. During the tests subtropical or tropical conditions are frequently imitated, by placing specimens in a weatherometer or in a moist chamber heated to 40-60°C.

During tests in a moist atmosphere attention is by no means always paid to the geometrical factor, *ie* to the relationship between the free surface area of the metal and that in contact with the rubber. When metal partially screened by rubber is kept in atmospheric conditions, two contrasting zones are formed on it - a cathode one and an anode one; hence the direction and development of the corrosive process will be dependent on the relationship between surface areas of the metal and the rubber⁵²⁻⁵⁴.

After the tests and careful examination of the rubber/metal specimens before the gravimetric measurements an additional study is sometimes made of the corroded surface of the metal by means, for example, of a binocular Linnik microscope⁵, and also of an electron microscope^{11,55}. For the metallographic examination of the slides a metallo-microscope is used, for example, a MM-8 type³⁸. The corrosion effects are frequently determined by the electrical resistance method⁵⁶⁻⁵⁸ or by means of electro-chemical measurements, using additional or somewhat modified methods and instruments⁵. The surface of a metal corroded to a slight extent is investigated by optical methods, using a reflectometer^{59-61;62} or a goniometer⁶³ which make it possible to carry out precision measurements. The optical methods of investigation include ellipso-metrical and interference methods⁶⁴⁻⁶⁵.

The mechanism of the process of corrosion occurring between the polymer and the metal is studied by performing chemical and X-ray analysis⁶⁵⁻⁶⁹, and taking infra-red spectrograms and so on^{11,55}. For this purpose, impedance⁷⁰ and isotope⁷¹ methods may also be used, as well as the method of ion-ion emission⁷² which makes it possible to judge both the condition and the composition of the

surface layer of the metal from the character of the mass spectra of the secondary ions ejected by bombarding the primary ions.

The most reliable way of combating the corrosion of metals which are in contact with rubbers consists in the creation of particularly pure and stable rubbers which do not show corrosive activity in all recommended operating regimes. However, the difference in the cost, for example of ordinary butadiene nitrile rubber and a corrosively non-active one may be just as significant as that between ordinary and medical silicone rubbers. In accordance with the data in the literature⁴⁶, there is a complete absence of corrosive activity in terminated 822 and 823 X2 nitrile rubber where the acrylonitrile content is 31-34 weight per cent and the Mooney viscosity is 47. Rubber mixes based on it also have a reduced hygroscopicity. According to information from Naugatuck Chemical, USA, butadiene nitrile rubbers, Paracril BLT, BJLT, prepared by low-temperature polymerisation, do not cause corrosion of metals and are suitable for the preparation of rubbers with minimal corrosive activity²⁵. When using industrial polymers, which are inclined to produce corrosive materials, the mixes frequently include fillers which neutralise or reduce their corrosive activity to a permissible minimum. In chloroprene rubbers the vulcanising agents - oxides of zinc and magnesium, also play the part of acceptors of hydrogen chloride which is evolved during the storage and use of components¹³. In a number of cases positive results may be obtained by preliminary treatment of the rubbers with the object of removing volatile admixtures which are responsible for the corrosion of metals. This treatment may consist in heating or evacuation of polymer materials at ordinary or elevated temperatures, sometimes in combination with mechanical action.

In order to reduce the corrosive activity of SKN-18 + SKN-26 and SKN-18 + SKF-32 rubbers it is recommended that a corrosion inhibitor of the amine type - mercapto benzimidazole^{38,73} or bis(furylidene)hexamethylenediamine⁷⁴ should be used. It is possible to prevent corrosion of carbon steels which are in contact with fluoro-rubber SKF-32, which is susceptible to thermo-oxidation and hydrolytic attack, by introducing into the rubber mix one part by weight of dibutyl tin dilaurate⁷⁵.

A report has appeared about a new corrosion inhibitor bearing the trade mark 'Cornox'^{76,77}. This product is produced in the USA in the form of four modifications. Chromates are used in order to prevent corrosion of aluminium components which are in contact with sealants^{6,78}. Among the most accessible means for the protection of metals in contact with rubber are thin layer coverings of the paint and varnish type of lubricants. They are applied to the rubber and

create a barrier between the metal and the rubber and partly improve the sealing properties of the joints. Pure vaseline, petrolatum, and gun oil were tested as lubricants, preventing, in a 0.5n solution of NaCl, the corrosion of stainless steels in contact with rubbers of SKB, SKMC-30, SKT and so on. Somewhat worse results were obtained when gun oil was used⁵.

The rate of corrosion of metals in a limited space, for example in a closed housing can sometimes be reduced by varying the composition of the ambient atmosphere. In the most simple case, this can be attained by periodic ventilation of the closed volume or by drying the air enclosed in it by the well-known solid desiccants. A lowering of the temperature also retards the corrosive process in rubber/metal components, which has been noted during their long-term storage in warehouses.

The volatile products which are given off from rubbers, sealants or rubber coatings are on the whole acidic. Therefore it is possible to retard the process of corrosion by selecting the corresponding neutralising, volatile inhibitors which can be placed for example in the housing of the instrument with the rubber components. Among the passive, but most reliable methods of combating corrosion is the correct choice of metals which in practice do not corrode in contact with rubbers and sealants. It is well-known that the Soviet Thiokol sealant U-30M causes corrosion of copper, silver and their alloys, whereas ferrous metals and in particular chrome nickel alloys in this case are corrosion resistant²¹. In some cases, particularly with considerable corrosion, protective metallic or non-metallic coverings can be applied to the components^{79,80}. In real operating conditions of course combined methods of protection of metals against corrosion are used.

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